

Softening of the Bond-Stretching Phonon of $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ at the Metal to Insulator Transition

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The dispersion of phonons in $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ along the $(3+q, 0, 0)$ direction in reciprocal space was determined for $x=0, 0.30, 0.37, 0.52$ using high-resolution inelastic X-ray scattering. The observed phonon energies near Γ were in good agreement with published optical data. It was found that two high-energy modes strongly soften near $q=0.25$ when the system becomes metallic ($x > 0.35$). There was no softening in the insulating phase, even at $x=0.30$, very near the metal-insulator transition. This suggests that the phonon softening is linked to the appearance of the Fermi-surface.

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I. INTRODUCTION

The electron-phonon interaction plays a central role in conventional BCS-Eliashberg superconductors. While, in most cases, the phonons contributing to superconductivity are low frequency modes, there are two examples of superconductors in which a high frequency optical mode predominantly contributes to pairing: $\text{Ba}(\text{Pb,Bi})\text{O}_3$ with the superconducting transition temperature, $T_c=11$ K¹ and $(\text{Ba,K})\text{BiO}_3$ with $T_c=30$ K². In these materials the Bi-O breathing mode phonon strongly couples to the electronic system^{3,4}. Another example is MgB_2 , $T_c=39$ K⁵, where the coupling to the high-energy boron optical mode plays a main role in superconductivity⁶.

Strong electron-phonon coupling often manifests itself as phonon softening. For MgB_2 , inelastic X-ray scattering (IXS) has demonstrated that the dispersion of the E_{2g} phonon strongly softens and broadens near zone center^{7,8}. Subsequent work with carbon substituted MgB_2 revealed that the amount of phonon softening correlates with T_c ⁹, providing direct proof that this phonon is responsible for the high T_c superconductivity in MgB_2 .

By contrast, in the high- T_c superconducting cuprates (HTSCs), the phonon contribution to superconductivity remains unclear, although the high-frequency oxygen mode (Cu-O bond stretching mode) shows strong softening as the momentum transfer is changed¹⁰⁻¹⁷. The main subject of discussion for the cuprates is the rapid drop of the bond stretching phonon frequency, and increased linewidth, in the $(q, 0, 0)$ direction at $q > 0.25$, which is not reproduced by the LDA calculations^{14,18,19}. The carrier doping dependence of the phonon softening is also an unresolved problem. Namely, why the softening gradually develops with hole doping and saturates at the high doping levels¹⁵ is not well understood.

The latter problem motivated us to investigate the doping dependence of phonon softening in other superconducting systems near a metal-insulator (MI) transi-

tion. Phonon softening near a MI transition is usually observed when temperature is lowered. However, there are few studies of phonon softening as a function of carrier doping by chemical substitution, particularly in system where there is a doping-induced MI transition. From this viewpoint, $(\text{Ba,K})\text{BiO}_3$ (BKBO) is a good candidate for study. BKBO has a perovskite structure, similar to the cuprates. As K is doped into the system, BKBO changes from a charge-density-wave (CDW) insulator to a metal that superconducts ($T_c \sim 30$ K) at $x \sim 0.35$. Crystal structure also changes with x from monoclinic to orthorhombic (at $x \sim 0.1$) and then to cubic structure (at $x \sim 0.35$)²⁰.

The phonon dispersion of BKBO has previously been investigated only by neutron scattering measurements for K concentrations of $x=0.02$ and 0.40 ^{21,22}. That work showed that no phonon softening was observed in the insulator, while dramatic softening of the Bi-O stretching and breathing modes appeared in the metallic (superconducting) phase. It is then interesting to consider how the softening changes as one increases x through the MI phase transition, for example, whether the phonon softens only near the MI transition or whether the phonon softening correlates with T_c as the electron-phonon coupling changes with K concentration. However, the earlier work^{21,22} was limited to only the two K concentrations.

In the present paper we investigate the phonon dispersion of BKBO single crystals with $x=0, 0.30, 0.37$, and 0.52 in order to clarify the phonon behavior near the MI transition. Despite great effort, we were not able to grow single-phase samples with dimension larger than about 0.5 mm, however, our technique of measuring phonon dispersion, IXS, allows us to perform these measurements on such small samples. The samples with $x=0$ and 0.30 are in the insulating phase, while those with $x=0.37$ and 0.52 are metallic. So far, there has been no report of phonon dispersion for $x=0.30$, near the MI transition, and for $x=0.52$, a heavily doped composition

of the metallic phase.

II. EXPERIMENTS

Single crystals of BaBiO_3 were grown by a flux method using Bi_2O_3 and BaCO_3 , and the K-doped crystals by the electrochemical technique using Bi_2O_3 , $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ and KOH ^{23–25}. Our sample sizes were typically $\sim 0.3 \times 0.3 \times 0.3 \text{ mm}^3$ ($< 0.2 \text{ mg}$) - while effort was made to find larger crystals, they were always multi-domain and/or non-uniform in doping. It was particularly difficult to grow homogenous samples with $x \sim 0.3$, and this may explain why there are relatively few experimental investigations of samples with $x \sim 0.3$ to 0.35. The lattice constants of the samples were measured using a four-circle X-ray diffractometer, and the K concentration was determined assuming a linear relationship between the lattice parameter and x in $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ ²⁰. Superconducting transition temperatures were determined from the magnetic susceptibility measured by a superconductor quantum interference device (SQUID) magnetometer. The K-content estimated from the lattice parameter was in good agreement with the value expected from the measured T_c ²⁰.

IXS spectra were measured at BL35XU of SPring-8²⁶. We measured spectra along (100) direction of the simple perovskite lattice, i.e. from Γ to X points, $(3+q, 0, 0)$, where $0 < q < 0.5$, at room temperature. Depending on setup, the energy resolution varied from 1.5 to 6 meV according to the choice of x-ray energy and analyzer Bragg reflection (1.5/3/6 meV resolution at the Si (11 11 11)/(999)/(888) at 21.7/17.8/15.8 keV). The incident beam at the sample position was about $60 \times 100 \mu\text{m}^2$ in the full-width at half maximum (FWHM). Because of the strong X-ray absorption by heavy elements, the X-ray penetration depth in BKBO is small ($< 20 \mu\text{m}$), for example, about 40% of that for $(\text{La}, \text{Sr})_2\text{CuO}_4$ which resulted in a relatively weak phonon signal. For the analysis of the IXS spectra, the pseudo-Voigt functions were used.

III. RESULTS

Figure 1 shows the IXS spectrum of BaBiO_3 at various q values. The spectra are well reproduced by fitting curves consisting of an elastic scattering peak and six phonon peaks. Since the measurement direction is $(3+q, 0, 0)$ in the present experiment, the intensity of longitudinal optical (LO) phonons is enhanced, while the transverse optical (TO) phonons are almost undetectable. Since the intensity at energy higher than 80 meV is very small, we consider the features in this energy range unreliable and ignore them. Based on the published optical data^{27,28}, we assign the six peaks from low to high frequency as the acoustic mode, external mode (relative displacement of Ba and BiO octahedron), Bi

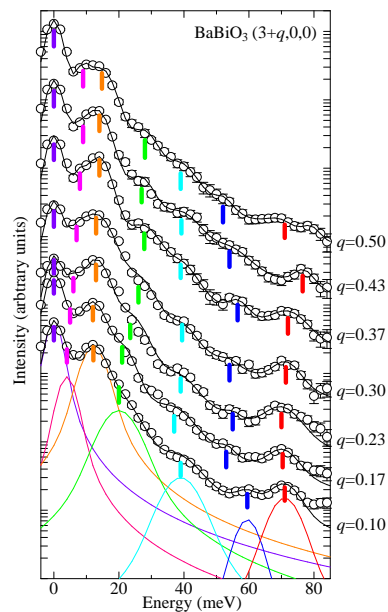


FIG. 1: (Color online) A typical IXS spectrum for BaBiO_3 system at various q values. The open circles show the experimental data with their statistical error bars. The lines below the spectrum show the pseudo-Voigt fitting for elastic scattering and each phonon mode indicated by a short bar. The black solid lines show the total fitting of the experimental data.

mode, Bi-O bending mode, Bi-O stretching mode, and Bi-O breathing mode. Although there are only four longitudinal modes, including the acoustic mode, for a cubic perovskite structure, the lattice distortion coupled with CDW creates additional phonon branches. For example, the Bi-O breathing mode at the zone boundary becomes a zone center mode that is Raman active. The Bi-Bi vibration mode that is originally an acoustic mode turns to an infrared active mode because of inequivalent neighboring Bi charges²⁷.

Figure 2 illustrates a comparison of our data near the Γ point ($q \sim 0.1$) with published optical data including both far-infrared and Raman scattering measurements^{27–31}. As seen in the figure, our IXS data agree well with the optical data over the whole composition range, except for the peak at 60 meV. This is strong support for the reliability of our measurement. As for the 60 meV peak, we tentatively attribute it to the LO bond-stretching mode, although its energy is different from the far-infrared data (69 meV). Another possibility might be a TO-mode, which, while it should be weak in our IXS data, is known to be at 60 meV from far-infrared measurements.

We now focus on the higher frequency phonons. In Fig. 3, the high-energy part of the spectra ($\hbar\omega > 45 \text{ meV}$) is shown at several q -values for all of the measured crystals. For the insulating samples with $x=0$ and 0.30, the q -dependence of the phonon frequency is weak, while the phonons in the metallic samples with $x=0.37$ and 0.52 exhibit large changes between the second and the

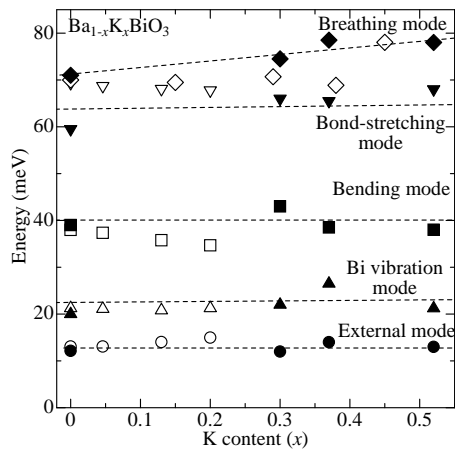


FIG. 2: The potassium content dependence of phonon energies for various modes at $q \sim 0.1$ in $(3+q,0,0)$ momentum (closed symbols). Open symbols are the other experimental data for the LO phonon modes from Raman and optical spectroscopies^{28–30}. The dashed lines are guide for eyes.

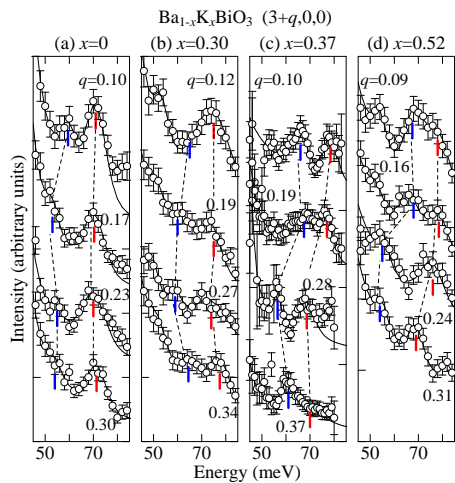


FIG. 3: (Color online) High energy part of IXS spectrum of BKBO system for the four compositions. The number beside the data shows the q value in $(3+q,0,0)$ momentum. The open symbols show the experimental data and the solid lines show the fitting result. The short bars on the data indicate the positions of the phonon peaks. The vertical dashed lines are guide for eyes to track the phonon dispersions. The data were plotted by a shift in the vertical axis to distinguish the different q patterns.

third q vectors (at $q \sim 0.2$).

The observed phonon frequencies at all the q -values are summarized in Fig.4. Anomalous softening of the bond stretching and the breathing modes is observed for $x=0.37$ and 0.52 , while almost no softening is seen for $x=0$ and 0.30 . The neutron scattering results of Braden^{21,22} are also plotted for comparison. The agreement with the neutron data is good for $x=0$, but not for $x=0.37$. This discrepancy between our data and the neutron result might be due to a difference in K-content

of the samples. Our IXS results are in better agreement with the optical data. Although the phonon frequencies are different, the tendency of phonon softening is the same in both neutron scattering and IXS experiments.

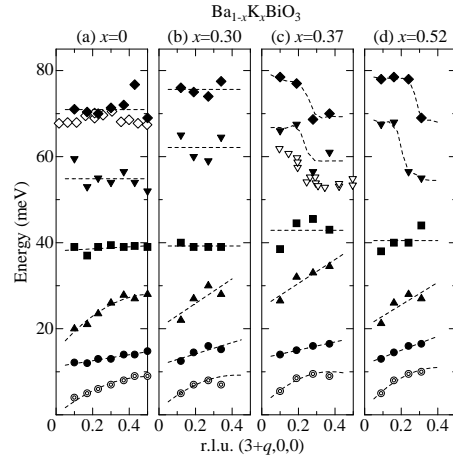


FIG. 4: Phonon branches of BKBO system in (100) direction for four different values of x (closed symbols and double circles) and the data from neutron scattering study^{21,22} for comparison (open symbols). The dashed lines are guides for eyes.

It should be noted that the breathing mode as well as the Bi-Bi vibration mode survives even in the metallic phase, although these modes should not be present in a cubic perovskite structure without the CDW distortion. The present results suggest that local lattice distortions remain even in the nominally cubic phase. In fact, we observed strong superlattice peaks during the experiments, as if the CDW distortion remained. Also there have been several reports on the short range structure distortion suggested by X-ray absorption fine structure³², ultrasonic measurements³³ and Raman scattering³⁴ in the metallic phase. Recently, even a neutron diffraction measurement has detected a long-range structural distortion in the superconducting BKBO characterized by a tilt of the BiO octahedra³⁵.

IV. DISCUSSIONS

An important result of the present study is that no softening is observed in the phonon dispersion for $x=0.3$, very close to the MI transition in the insulator phase. Often (in a conventional CDW material, for example), phonon softening occurs as the temperature is swept through the metal-insulator transition, with the softening being largest at the phase transition, and then recovering. In the present case, we change the doping at a fixed temperature (300K), and the crystal structure changes from orthorhombic to cubic at $x \sim 0.35$, accompanied by the change from a CDW insulator to a metal. (The change from monoclinic to orthorhombic at $x=0.1$ does not seem

to affect the phonon dispersion.) Making an analogy between T - and x -sweep experiments, we expected phonon softening at x just before and after the MI transition. The absence of the phonon softening at $x=0.3$ may be because the MI transition is too sharp as a function of x (or T) to follow the transient behavior of the phonon. Since even near $x=0.35$ the MI transition temperature is much higher than room temperature, we cannot observe softening at 300K. This fact implies that the CDW gap is still large at $x=0.30$ as was suggested optical absorption data ($\sim 0.5\text{eV}$ at $x=0.28$, while 2eV at $x=0$)³⁶.

In contrast, clear phonon softening in BKBO is observed in the metallic phase with a large Fermi surface (FS). Once the FS is formed after the phase transition, it changes only weakly with x , which might result in the x -independent ($x > 0.35$) softening seen in the present work.

According to the theoretical prediction, the electron-phonon coupling constant, λ , which has the largest contribution from the breathing mode, decreases with increasing x ⁴, because the FS shrinks, as the band-filling is reduced. This can explain why T_c for $x=0.52$ is lower than that for $x=0.37$. However, the present results do not show a clear x - (or T_c -) dependence of phonon softening in the metallic phase. Therefore, at least in the present case, the amount of softening of the bond stretching phonon is not a good measure for the strength of the electron-phonon coupling that determines T_c .

The reason for this unexpected result is not clear. One possibility is that the phonon change is too small to be observed with the present experimental resolution. According to the Shirai's calculation⁴, λ changes from 0.88 ($x=0.4$) to 0.53 ($x=0.5$), from which T_c was estimated as 25K ($x=0.4$) and 10K ($x=0.5$), on the basis of the Allen-Dynes equation³⁷. These values are not far from the experimental data (30 K for $x=0.4$ and 17 K for $x=0.5$)²⁰. Compared to such a drastic change in T_c with λ , the phonon frequency is expected to show a weaker λ -dependence. The phonon renormalization due to electron-phonon coupling is generalized as $\omega_{ph}^2 \sim \Omega_{ph}^2 / (1 + \lambda)$ ³⁸, where ω_{ph} is a renormalized phonon frequency and Ω_{ph} is a bare phonon frequency³⁹. This expression gives about 10 % change in phonon frequency when λ changes from 0.88 ($x=0.4$) to 0.53 ($x=0.5$), which is much smaller than the 60 % reduction in T_c . While the above relation between λ and the renormalized phonon frequency is a very rough approximation, the important idea in the present context is that the fractional changes in phonon frequency may be small compared to changes in λ .

Finally, we note that the present result brings the unusual phonon behavior of the HTSCs into sharp relief. Softening of the bond stretching phonon has

been reported in cuprates with the layered perovskite structure.^{10–17}, however, the doping dependence in the cuprates¹⁵ is very different. In BKBO, the electronic change with doping is small in the metallic phase, where a rigid band picture is applicable as a first approximation. In this situation, as we see in the present study, even near the MI transition, 10 % change of band filling⁴⁰ gives only a little change in phonon softening. By contrast, in the underdoped cuprates (for example, at $0 < x < 0.15$ of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$), the system is in the so-called "pseudogap" state⁴¹ where the FS rapidly develops with carrier doping from a Mott insulator⁴². Therefore, it is likely that such a radical change of FS topology accompanied with a huge enhancement of the electronic density of states is responsible for the gradual increase in softening of the Cu-O bond stretching phonon with carrier doping. The importance of FS topology in electron-phonon coupling of the cuprates was also pointed out in ref.¹⁷. Further detailed information about FS will promote more precise calculations of the phonon behaviors in the cuprates.

V. SUMMARY

We have investigated the phonon dispersion of $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ in the $(3+q, 0, 0)$ direction for $x=0, 0.30, 0.37$, and 0.52 using IXS. In contrast to the nearly doping independent behavior of the low frequency modes, the higher frequency modes related to Bi-O bond stretching vibrations showed a clear and abrupt change with K-content. In the insulating phase, $x < 0.35$, mode dispersions were almost flat, while radical softening was observed towards zone boundary in the metallic phase, $x > 0.35$. The degree of softening is relatively unaffected by K-content ($x > 0.35$). This doping dependence of the phonon softening suggests that the present softening is different than the softening often associated with a structural phase transition, and, more probably, is electronic in origin.

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 - ⁴⁰ In a rigid band picture with a half-filled band at $x=0$ and zero filling at $x=1$, the change in x from 0.37 to 0.52 corresponds to the 7.5 % decrease in band filling. This gives only 2.6 % reduction in Fermi wavenumber (radius of Fermi sphere).
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